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# EUROPEAN PATENT APPLICATION

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54 Production of high concentration tocopherols and tocotrienols from palm oil by-products.

57 The invention relates to a process for the production of tocopherols (T) and tocotrienols (T3) from palm fatty acid distillates (PFAD). The process comprises of converting free fatty acids and glycerides in PFAD into alleged esters, then separating T and T3 from the alkyl esters and other impurities. The T and T3 is concentrated by ion-exchange and further concentrated by distilling the resulting product. Specific catalysts and optimum temperatures for the process are disclosed.

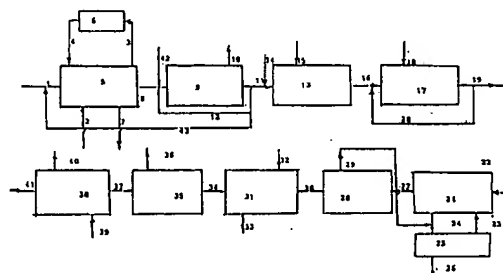


Figure 1

## Description

**PRODUCTION OF HIGH CONCENTRATION TOCOPHEROLS AND TOCOTRIENOLS FROM PALM OIL BY-PRODUCT**

The present invention relates to a novel method for the production of tocopherols (T) and tocotrienols (T3) from palm oil by-product such as Palm Fatty Acid Distillate (PFAD).

Tocopherols and tocotrienols are very useful substances exhibiting strong antioxidant activities and physiological activities. High concentrates of tocopherols and tocotrienols are not easily obtained by concentration of PFAD because the amounts of tocopherols and tocotrienols in PFAD are very low compared to soyabean, rapeseed and similar raw materials. PFAD is composed mainly of fatty acids, sterols, tocopherols, tocotrienols, squalene and like impurities.

Known processes for the concentration of tocopherols and tocotrienols usually use solvent extraction, solvent fractionation, ion-exchange resin treatment, etc., at the laboratory stage, but these processes are not complete or economically attractive. The present invention seeks to provide a combination of unit processes which produce better quality and better yield compared to the previous proposals. PFAD relatively contains high level of tocotrienols compared with other sources and this has not been commercially exploited. It is therefore an object of the present invention to provide a novel and efficient method for the production of tocopherols and tocotrienols from PFAD.

According to one aspect of the present invention, there is provided a process for the production of tocopherols (T) and tocotrienols (T3) from palm fatty acid distillates (PFAD) which comprises:

- converting free fatty acids and glycerides in PFAD into alkyl esters;
- separating T and T3 from the alkyl esters and other impurities;
- concentrating the T and T3 by ion-exchange; and
- distilling the resulting product to produce a further concentrated T and T3 fraction.

More specifically in accordance with the present invention, there is provided a process for production of tocopherols and tocotrienols from palm fatty acid distillates (PFAD) which comprises:

- (a) treating the PFAD with an alkyl alcohol and appropriate catalysts to convert free fatty acids and glycerides into alkyl esters by esterification and transesterification, respectively;
- (b) distilling the resulting product under reduced pressure to remove a major part of the alkyl esters and leave the tocopherols, tocotrienols (T and T3) and other higher boiling point substance in the residue;
- (c) cooling the residue to bring about crystallization of higher melting point substances and other impurities and filtering off the crystalline material to leave the T and T3 in the filtrate;
- (d) treating the filtrate from (c) by an ion-exchange procedure with a high selectivity anionic resin to produce a concentrated T and T3 fraction;
- (e) removing the solvent from the T and T3 fraction from (d) by evaporation;
- (f) washing and drying the product from step (e);
- (g) subjecting the product from (f) to molecular distillation to produce a further concentrated T and T3 product;
- (h) deodorising the T and T3 product.

In a modified form of the above process, the PFAD is pretreated (before esterification) by distillation to remove a major part of the free fatty acids.

By optimizing the conditions for the various steps described above, it is possible to produce a product having a high concentration of tocopherols and tocotrienols with very low losses of material during the process.

A discussion of the preferred conditions for the steps described above follows:

- (a) It is preferred to use p-toluenesulfonic acid, hydrochloric acid or sulphuric acid as the catalyst for conversion of free fatty acid in PFAD into alkyl esters, at temperatures between 65° and 110°C and reaction times of less than 3 hours.

Potassium hydroxide, sodium hydroxide or sodium methoxide are preferred as catalysts for conversion of glycerides into alkyl esters at temperature between 30° and 70° C with reaction times of 10 minutes or more.

It is also preferred to treat the reaction mixture with a chelating agent such as ascorbic acid (Vitamin C), phosphoric acid, maleic acid, citric acid or tartaric acid, before drying and distillation.

- (b) Distillation is preferably carried out using a high heat-transfer rate falling film vacuum distillation column, operating at below 10 torr (1333 N/m<sup>2</sup>) and at a temperature between 100° and 200°C. Under such conditions, it is possible to concentrate T and T3 from 0.5% to more than 10%, with losses of T and T3 in the distilled alkyl esters being less than 1% based on the original raw material.

- (c) It is preferred to concentrate T and T3 by using an anion-exchange resin column using methanol, ethanol or hexane as the eluting solvent and an acidic solution, such as sulphuric acid or boric acid, for desorbing T and T3 from the ion-exchange resin. Concentration from an initial 8% up to 80% or more can be achieved in this manner.

(e) Solvent evaporation is preferably carried out using a falling film evaporator and a rotary short path evaporator in series operating at 50°C and 130°C respectively, and under reduced pressure which minimises denaturation of tocopherol and tocotrienols.

(g) It is preferred to carry out the molecular distillation at 140° to 220° C under a vacuum below 0.05 torr (6.7 N/m<sup>2</sup>). A T and T3 fraction with greater than 95% concentration can then be produced from raw materials containing a 60% concentration of T and T3.

The optional pretreatment (distillation) step is preferably carried out using a high heat-transfer rate falling film distillation column at temperature between 150° and 250° C and a vacuum below 10 torr. (1333 N/m<sup>2</sup>). 5

It is also preferred to minimise contact of tocopherols and tocotrienols with oxygen by nitrogen and/or nitrogen sparging throughout the various unit processes.

The process of the invention is further illustrated by the block flow diagram shown in Figure 1. Melted PFAD is fed into reaction vessel 5 via pipe system 1. A mixture of an alkyl alcohol and an acidic catalyst, such as p-toluenesulfonic acid (PTS) hydrochloric acid (HCl) or sulfuric acid, is introduced via pipe system 2. The reactants are heated and the esterification reaction is conducted at temperature between 65 and 110° C. Alkyl alcohol is continuously introduced into reaction vessel 5 via pipe system 4 and the evaporated alkyl alcohol is recovered and purified by the condensation and distillation set-up 6 via pipe system 3. When the reaction is completed, the reaction mixture is cooled. Another mixture of an alkyl alcohol and a catalyst, such as potassium hydroxide, sodium hydroxide or sodium methoxide, is added into reaction vessel 5 via pipe system 2 and transesterification of glycerides proceeds at temperature between 30° and 70° C and at reaction time of 10 minutes or more. After treatment with chelating chemical such as vitamin C, phosphoric acid, maleic acid, citric acid and tartaric acid, water washing, nitrogen sparging and drying, the resulting product is passed to distillation equipment 9 via pipe system 8. Effluent is discharged via pipe system 7. 10 15

Distillation equipment 9 consists of high heat-transfer distillation column and distillate collection system. Distillation process is continuous. Alkyl esters are distilled at high vacuum at below 10 mm of Hg and at temperature between 100-200° C. Distilled alkyl esters are collected by condensation and discharged via pipe system 10 as a by-product. Retention time of T-T3 in the distillation column is short that deterioration is minimum. More than one distillation cycle may be practiced and recycling of the heavy phase is by pipe system 12. The final heavy phase of distillation equipment 9 is a mixture of T&T3 and other substances found in PFAD and is passed to crystallizer 13 via pipe system 11. 20 25

The mixture in crystallizer is heated to homogeneous and then cooled to 0-15° C in between 5-30 hours of cooling time by a programmable automatic control system. Various quantities of solvent such as acetone, ethanol and methanol, may or may not be added into the mixture in crystallizer 13 before cooling started via pipe system 14. 0.5% or more of filter aid are added into the crystallizer via pipe system 15. 30

The mixture in crystallizer 13 is then passed to filter 17 where the crystallized substances are retained in filter cakes via pipe system 16. Before filtration started recycling or filtrate via pipe system 20 may be practiced in order to form sufficient cake thickness on filter element. Positive pressure filtration is practiced either by using pump or by applying nitrogen gas via pipe system 18. The final filtrate which is basically free of higher melting point substances is passed to ion-exchange process set-up 21 via pipe system 19. 35

The filtrate is introduced into an ion-exchange column which consists of regenerated anion resin packing with high selectivity in adsorbing T-T3. Acidic solution such as sulfuric acid or boric acid is used to desorb T-T3 from the anion resin via pipe system 22. Solvent such as methanol, ethanol and hexane, is used for elution of the various fraction in the ion-exchange process coming from solvent recovery set-up 25 via pipe system 23. Undesirable eluted fractions or effluent are discharged for solvent recovery or for other processing via pipe system 24 and 26 respectively while the desired fractions which contain reasonable high concentration of T-T3 are passed to evaporation equipments 28 via pipe system 27. 40

The evaporation system 28 is designed to provide short retention time for the T-T3 concentrate under vacuum that deterioration of T-T3 is minimum. Evaporated solvent is condensed and sent for purification via pipe system 29. The solvent free T-T3 concentrate is passed to washing and drying equipments 31 via pipe system 30. 45

Water is added into a mixing vessel containing the T&T3 concentrate from evaporation equipment 28 via pipe system 32. Mixing or washing are conducted at elevated temperature under nitrogen blanket. Effluent is discharged via pipe system 33. Drying is carried out in the same or different equipment by vacuum at temperature between 90-100° C. The resulting washed and dried product is passed to molecular distillation equipment 35 via pipe system 34. 50

Molecular distillation is carried out at very high vacuum. High concentration of T&T3 fraction is obtained, at temperature between 140-220° C and at vacuum below 0.05 torr 6.67 N/m<sup>2</sup>. Undesirable fractions are discharged via pipe systems 36 while the high concentration T&T3 fraction is passed to deodorization equipment 38 via pipe system 37. 55

Deodorization is conducted at temperature between 180-250° C and at vacuum of 3-15 torr (400-2000 N/m<sup>2</sup>). Low pressure steam is introduced into the deodorizer via pipe system 39 at a rate of 1-6% of the treated material. Distillate is collected by condenser via pipe system 40. Odourless final high concentration T&T3 is sent for consumption via pipe system 41. 60

All the process equipments described in above are equipped with nitrogen gas blanketing and nitrogen vacuum break systems for protecting T-T3 from oxidation.

## Process 2

This process is similar to that of Process 1 except that the raw material, PFAD is pretreated by removing majority of the free fatty acid in PFAD by distillation before sending for processing by Process 1 at the same 65

sequencing as described from (a) to (h) at Process 1.

As illustrated by the same Figure as Process 1, PFAD is passed to the storage facility of distillation equipment 9 via pipe system 42. Distillation process is continuous. Majority of the free fatty acids are distilled at high vacuum at below 10 torr (1335 N/m<sup>2</sup>) and at temperature in between 150-250°C. Distilled fatty acids are collected by condensation and discharged via pipe system 10. More than one distillation cycle may be practiced and recycling is via pipe system 12. The final heavy phase is passed to reaction vessel 5 via pipe system 43. The processing conditions and products flow from reaction vessel 5 onward are same as the described in Process 1.

## 10 Examples

### Process-1

Several experiments on process-1 were conducted. Every step's conditions of example-1 and example-2 are shown in Table 1.2 - 1.9, and the results are shown in Table 2.

During methylesterification MeOH was fed continuously into the reaction vessel. And water which was by-produced during methylesterification was removed continuously for effective reaction. Acid Value decreased below 0.1 after methylesterification. Almost all glycerides which are contained in the sample (about 10%) were transesterified with catalyst. Ascorbic acid solution was used for protection of T & T3 from denaturation. Samples were dried until the moisture was less than 0.5%

Fatty acid methylesters were removed by distillation and this treatment could achieve more than 16 folds concentration in view of T and T-T3 concentration in heavy phase as shown in Table 2.

Impurities which have higher melting points than T & T3 in the heavy phase were removed by crystallization and filtration. Crystals appeared when the samples were cooled down with or without several kinds of solvent. Crystals were removed by filtration by using the pressure of nitrogen gas.

The filtrates were loaded to regenerated anion-exchange resin in a column. Then the column was washed with 95% EtOH to purge impurities which did not attach to the ion-exchange resin. 10% of acid solution was used to desorb T & T3 from the resin. And then detached T and T-3s were collected as T and T-3 fraction by using 96% of EtOH.

Evaporations were conducted in 2 steps under the conditions described in Table 1.6. During 1st step, mainly EtOH was evaporated and in 2nd step, solvents including water were evaporated completely. After evaporation of EtOH and water, the concentrations of T and T3 were 83.2%, 87.6% respectively as shown in Table 2.

Several batches of T and T3 were mixed and water-soluble impurities could be got rid of the sample by two times of washing with water, followed by drying under the vacuum.

Further purification of T and T3 could be achieved by molecular distillations which were conducted in 2 steps. Firstly at lower temperature, impurities which are easy to be evaporated could be removed and secondly at higher temperature T and T3 could be evaporated. The impurities which show higher boiling points than T and T3 remained in the heavy phase.

Steam deodorization after molecular distillation produced final product which had no smell and light-brown in colour.

Through our process above mentioned slight or no denaturation of T and T3 could be observed.

### Process-2

Several experiments on process 2 were conducted. Every step's condition of example-3 and example-4 are shown in Tables 1.1 - 1.9, and the results are shown in Table 2. Compared to above process-1, initial stage is different. First free acids in PFAD were distilled roughly before methylesterification in order to decrease the quantity of material which is to be methylesterified. The succession of other treatments after this distillation is the same as process-1.

Table 1.1  
Conditions of fatty-acid distillation

Factor		Ex-1	Ex-2	Ex-3	Ex-4
Vacuum	(toor)	-	-	1.5	2.0
Temp.	(c)	-	-	185	195
Time	(hr)	-	-	5.0	5.0

Table 1.2

## Conditions of methylesterification

Step & Factor	Ex-1	Ex-2	Ex-3	Ex-4	
Esterification					
Catalyst,	H <sub>2</sub> SO <sub>4</sub> 0.2%	H <sub>2</sub> SO <sub>4</sub> 0.15%	H <sub>2</sub> SO <sub>4</sub> 0.05%	H <sub>2</sub> SO <sub>4</sub> 0.02%	
Temp. (c)	90 c	95 c	90 c	95 c	
Time (hr)	3	4	1.5	1.5	10
MeOH feed (l/h)	30	40	30	40	
Transesterification					
Catalyst,	Ca(OH) 0.5%	NaOH 0.4%	NaOH 0.5%	KOH 0.5%	
Temp. (c)	50 c	55 c	55 c	55 c	15
Time (hr)	2.0	1.5	2.5	2.5	
Chelator	Ascorbic-acid	Ascorbic-acid	Ascorbic-acid	Ascorbic-acid	

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Table 1.3

## Conditions of methylester-distillation

Factor		Ex-1	Ex-2	Ex-3	Ex-4	
Vacuum	(torr)	1.5	2.0	1.0	1.0	
	(N/m <sup>2</sup> )	(200)	(267)	(133)	(133)	
Temp.	(c)	150	150	180	180	30
Time	(hr)	8	10	4.5	5.0	

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Table 1.4

## Conditions of Crystallization and filtration

Step & Factor	Ex-1	Ex-2	Ex-3	Ex-4	
Crystallization					
Solvent	EtOH 20 1	No addition	MeOH 20 1	Hexane20 1	
Cooling temp. (c)	0 c	10 c	0 c	0 c	
Cooling time (hr)	24	24	24	24	45
Filtration					
Filtration aid (%)	3.0	2.0	4.0	4.0	
Filtration-pressure (kg/cm)	7	7	7	7	50

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Table 1.5  
Conditions of ion-exchange column

Solvent	Ex-1	Ex-2	Ex-3	Ex-4
Purging (1)	95% EtOH 40	95% EtOH 30	95% EtOH 40	95% EtOH 30
Detaching (1)	10% Boric acid 10	10% Formic acid 10	10% Lactic acid 10	10% Malic acid 10
Elution (1)	99% EtOH 60	99% EtOH 40	99% EtOH 50	99% EtOH 50

Table 1.6  
Conditions of evaporation

Step & Factor	Ex-1	Ex-2	Ex-3	Ex-4
1st step				
(N/m <sup>2</sup> )	(2666)	(2666)	(2666)	(2666)
Vacuum (torr)	20	20	20	20
Temp. (c)	80	80	80	80
Time (hr)	4	4	4	4
2nd Step				
(N/m <sup>2</sup> )	(267)	(267)	(267)	(267)
Vacuum (torr)	2	2	2	2
Temp. (c)	100	100	100	100
Time (hr)	1.5	2.0	1.5	1.5

Table 1.7  
Condition of washing & drying

Step & Factor	Ex-1	Ex-2	Ex-3	Ex-4
1st washing				
Water (l, c)	40,60	40,60	40,70	40,70
Stirring (min.)	20	20	20	20
2nd washing				
Water (l, c)	20,60	20,60	20,70	20,70
Stirring (min)	20	20	20	20
Drying				
(N/m <sup>2</sup> )	(400)	(533)	(400)	(533)
Vacuum (torr)	3	4	3	4
Temp. (c)	95	95	90	100
Time (hr)	1.5	1.5	1.5	1.5

Table 1.8

Conditions of molecular distillation

Step & Factor		Ex-1	Ex-2	Ex-3	Ex-4	5
1st step						
Temp.	(c)	120	130	135	130	
Vacuum	(torr)	0.04	0.02	0.03	0.03	
	(N/m <sup>2</sup> )	(5.33)	(2.67)	(4.0)	(4.0)	10
2nd Step						
Temp.	(c)	140	170	220	200	
Vacuum	(torr)	0.003	0.002	0.003	0.00	
	(N/m <sup>2</sup> )	(0.4)	(0.27)	(0.4)		15

Table 1.9

Conditions of deodorization

Factor		Ex-1	Ex-2	Ex-3	Ex-4	20
Temp.	(c)	160	180	200	200	
Vacuum	(torr)	2	2	2	2	25
	(N/m <sup>2</sup> )	(267)				
Steam feed	(g/hr)	500	400	300	450	
Time	(hr)	1.5	1.5	1.5	1.5	30



Table 2

The concentrations and accumulated yield of T &amp; T3 on each step

5	Step	Ex-1		Ex-2		Ex-3		Ex-4	
		Conc. (%)	Yield (%)	Conc. (%)	Yield (%)	Conc. (%)	Yield (%)	Conc. (%)	Yield (%)
	MA- T <small>ERIAL</small>	0.5	100	0.4	100	0.4	100	0.4	100
10	FA. DISTILLA- TION	-	-	-	-	2.1	97	2.4	98
	M.ES- TERIFI- CATION	0.5	98	0.4	99	2.1	95	2.4	94
15	M.E. DISTILLA- TION	8.2	95	10.1	96	8.4	91	9.5	90
20	CRY- STALLY- ZATION AND FILTRA- TION	8.3	90	10.1	92	8.4	88	9.5	88
25	ION-EX- CHANGE AND EVAP- ORATION	83.2	85	87.6	80	85.1	78	93.4	78
30	WASH- ING AND DRYING	83.8	82	87.8	78	85.4	76	83.7	77
35	MOLE- CULAR DIST. DEO- DORIZA- TION	96.2	75	97.9	71	95.2	70	96.6	70
40									

Those skilled in the art will appreciate that the invention described herein is susceptible to variations and modifications other than those specifically described. It is to be understood that the invention includes all such variations and modifications which fall within its spirit and scope.

#### Claims

1. A process for the production of tocopherols (T) and tocotrienols (T3) from palm fatty acid distillates (PFAD) characterised in that it comprises:
  - converting free fatty acids and glycerides in PFAD into alkyl esters;
  - separating T and T3 from the alkyl esters and other impurities;
  - concentrating the T and T3 by ion-exchange; and
  - distilling the resulting product to produce a further concentrated T and T3 fraction.
2. A process for production of tocopherols and tocotrienols from palm fatty acid distillates (PFAD) as claimed in Claim 1, characterised in that it comprises the steps of:
  - (a) treating the PFAD with an alkyl alcohol and appropriate catalysts to convert free fatty acids and glycerides into alkyl esters by esterification and transesterification, respectively;
  - (b) distilling the resulting product under reduced pressure to remove a major part of the alkyl esters and leave the tocopherols, tocotrienols (T and T3) and other higher boiling point substance in the residue;
  - (c) cooling the residue to bring about crystallization of higher melting point substances and other impurities and filtering off the crystalline material to leave the T and T3 in the filtrate;
  - (d) treating the filtrate from (c) by an ion-exchange procedure with a high selectivity anionic resin to produce a concentrated T and T3 fraction;

- (e) removing the solvent from the T and T3 fraction from (d) by evaporation;
  - (f) washing and drying the product from step (e);
  - (g) subjecting the product from (f) to molecular distillation to produce a further concentrated T and T3 product;
  - (h) deodorising the T and T3 product.
3. A process as claimed in Claim 1 or Claim 2, characterised in that the PFAD is pretreated (before esterification) by distillation to remove a major part of the free fatty acids therefrom.
4. A process as claimed in Claim 2 or Claim 3, characterised in that, in step (a), p-toluenesulfonic acid, hydrochloric acid or sulphuric acid as the catalyst for conversion of free fatty acid in PFAD into alkyl esters, at temperatures between 65° and 110° C and reaction times of less than 3 hours.
5. A process as claimed in any one of Claims 2 to 4, characterised in that, in step (a), potassium hydroxide, sodium hydroxide or sodium methoxide are used as catalysts for conversion of glycerides into alkyl esters at temperature between 30° and 70° C, with reaction times of 10 minutes or more.
6. A process as claimed in any one of Claims 2 to 5, characterised in that, in step (a), the reaction mixture is treated with a chelating agent such as ascorbic acid (Vitamin C), phosphoric acid, maleic acid, citric acid or tartaric acid, before drying and distillation.
7. A process as claimed in any one of Claims 2 to 6, characterised in that, in step (b), distillation is carried out using a high heat-transfer rate falling film vacuum distillation column, operating at below 10 torr (1333 N/m<sup>2</sup>) and at a temperature between 100° and 200° C.
8. A process as claimed in any one of Claims 2 to 7, characterised in that, in step (d), T and T3 are concentrated by using an anion-exchange resin column using methanol, ethanol or hexane as the eluting solvent and an acidic solution for desorbing T and T3 from the ion-exchange resin.
9. A process as claimed in any one of Claims 2 to 8, characterised in that, in step (e), solvent evaporation is carried out using a falling film evaporator and a rotary short path evaporator in series operating at 50° C and 130° C respectively, and under reduced pressure.
9. A process as claimed in any one of Claims 2 to 8, characterised in that, in step (e), solvent evaporation is carried out using a falling film evaporator and a rotary short path evaporator in series operating at 50° C and 130° C respectively, and under reduced pressure.
10. A process as claimed in any one of Claims 2 to 9, characterised in that, in step (g), the molecular distillation is carried out at 140° to 220° C under a vacuum below 0.05 torr (6.7 N/m<sup>2</sup>).
11. A process as claimed in Claim 3, characterised in that, the optional pretreatment (distillation) step is carried out using a high heat-transfer rate falling film distillation column at temperature between 150° and 250° C and a vacuum below 10 torr. (1333 N/m<sup>2</sup>).
12. A process as claimed in any one of the preceding Claims, characterised in that in order to minimise contact of tocopherols and tocotrienols with oxygen, nitrogen and/or nitrogen sparging is used throughout the various unit processes.

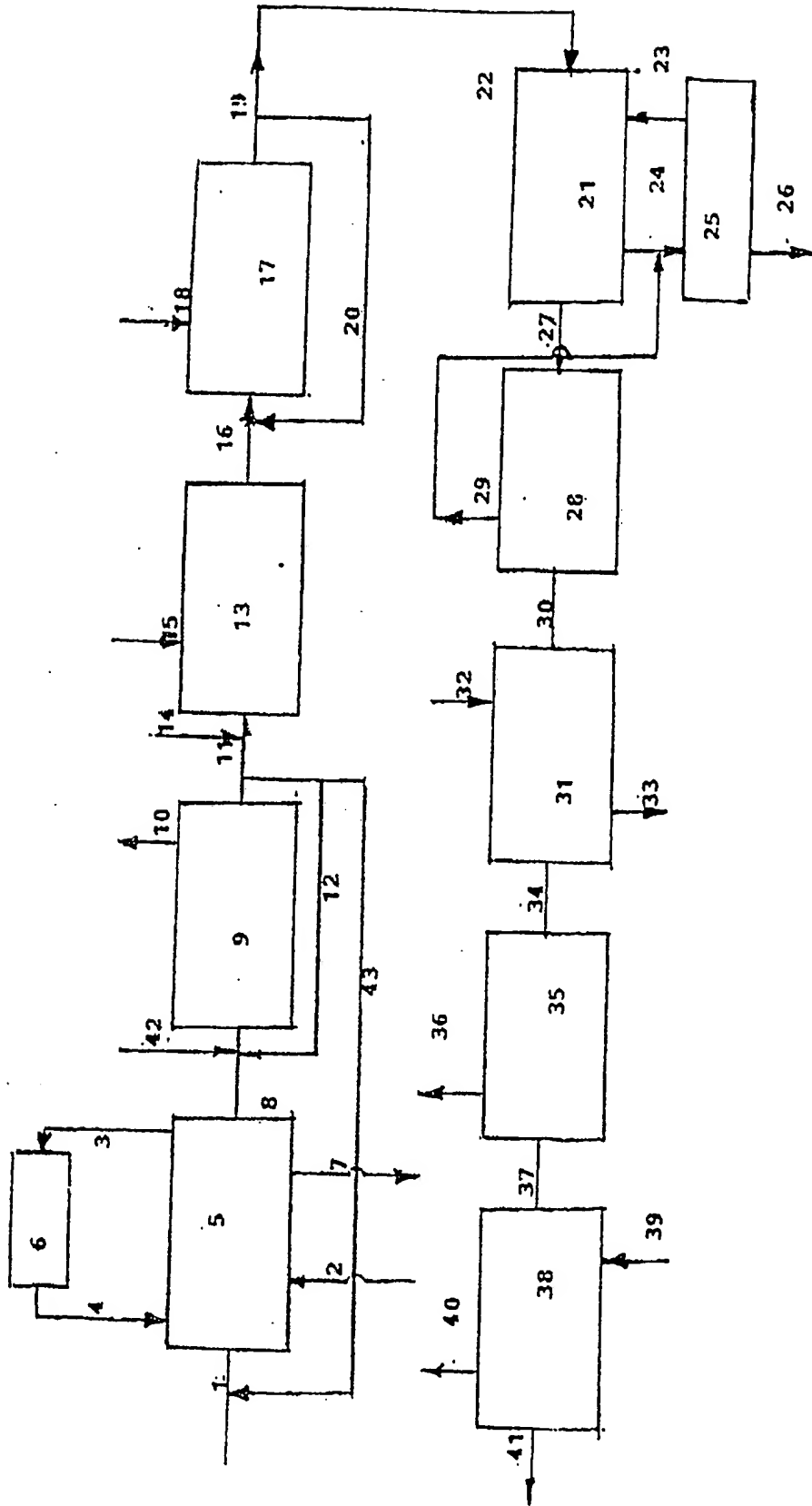


Figure 1

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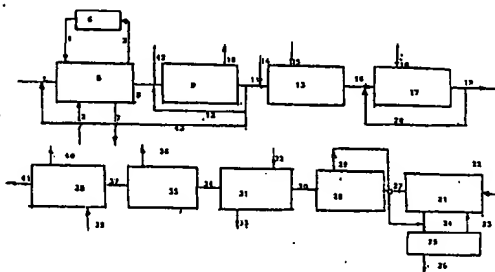
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**(54) Production of high concentration tocopherols and tocotrienols from palm oil by-products.**

57) The invention relates to a process for the production of tocopherols (T) and tocotrienols (T3) from palm fatty acid distillates (PFAD). The process comprises of converting free fatty acids and glycerides in PFAD into alleged esters, then separating T and T3 from the alkyl esters and other impurities. The T and T3 is concentrated by ion-exchange and further concentrated by distilling the resulting product. Specific catalysts and optimum temperatures for the process are disclosed.



### Fluorene 1



European Patent  
Office

# EUROPEAN SEARCH REPORT

Application Number

EP 89 30 2597

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl. 4)
X,Y	PATENT ABSTRACTS OF JAPAN, vol. 10, no. 31 (C-327)[2088], 6th February 1986; & JP-A-60 185 776 (RIKEN VITAMIN OIL K.K.) 21-09-1985 * Abstract *	1-3,7,9,10	C 07 D 311/72 C 11 C 1/00
Y	PATENT ABSTRACTS OF JAPAN, vol. 10, no. 269 (C-372)[2325], 12th September 1986; & JP-A-61 93 178 (AGENCY OF IND SCIENCE & TECHNOL) 12-05-1986 * Abstract *	1-3	
Y	GB-A-2 090 836 (AGENCY OF INDUSTRIAL SCIENCE AND TECHNOLOGY) * Page 1-4 *	1-4,7-9	
A	US-A-4 454 329 (YOSHIKI TAKAGI) * Column 1-4; claims *	1	
			TECHNICAL FIELDS SEARCHED (Int. Cl. 4)
			C 07 D 311/00
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 22-09-1989	Examiner FRANCOIS J.C.L.
CATEGORY OF CITED DOCUMENTS			
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons ..... & : member of the same patent family, corresponding document			